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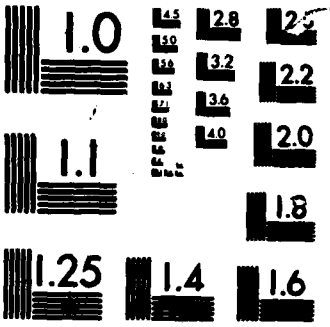

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DYNAMICS OF OSTWALD RIPENING WITH COALESCENCE

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ABSTRACT

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Numerical results of the Ostwald Ripening problem with coalescence are reported for aerosol with the continuum diffusive growth laws. It was shown that $N(t) \sim t^{-1}$, where $N(t)$ is the total number concentration. These results, though consistent with the asymptotic theory of Ostwald Ripening according to Lifshitz-Slyozov and with the similarity theory for the continuum Brownian coalescence process, show that the scaling arguments are insufficient to establish which of the two mechanisms is dominant asymptotically.

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Numerous studies have been concerned with the asymptotic stages of growth by condensation/evaporation of droplets or particles following their appearance by a homogeneous nucleation process. Comparatively less attention has been given the associated problem of growth when both condensation/evaporation and coalescence occur. For aerosols we present the first quantitative study of the Ostwald ripening problem with a realistic coalescence process.

We have investigated the isothermal spatially homogeneous growth of particles suspended in supersaturated monomer vapor and inert host gas. The process is described by the evolution equation for the singlet density function n(x,t):

$$\frac{\partial n(x,t)}{\partial t} + \frac{\partial}{\partial x}\left[\psi(x,s)n(x,t)\right] = \int_{x^*}^{x/2} b(x-x',x')n(x-x',t)n(x',t)dx' - n(x,t)\int_{x^*}^{\infty} b(x',x)n(x',t)dx' \qquad (1)$$

where $n(x,t)dx$ is the number of particles having masses in the range x,dx at time, t. $\psi(x,s)$ is the growth law for a particle by the condensation/evaporation process and $b(x',x)$ is the coalescence rate coefficient for two particles of masses x' and x. x^* is the mass of the smallest particle in the population and s is the supersaturation ratio of monomer vapor. We do not consider here competitive growth effects,[15,16] as the theory for this is lacking when coalescence is significant.

Equation (1) is coupled to the mass conservation equation for monomer vapor:

$$C_V \frac{ds}{dt} = -\int_{x^*}^{\infty} \psi(x,s)n(x,t)dx - x^*\psi(x^*,s)n(x^*,t) \qquad (2)$$

where C_V is the equilibrium bulk vapor concentration.

Equations (1) and (2) obey the mass conservation law:

$$\frac{d}{dt}(C_V s + M_1) = 0$$

where $M_1 = \int_{x^*}^{\infty} xn(x,t)dx$ is the mass concentration of particles. An important additional moment of $n(x,t)$ is the total number concentration, $N(t) = \int_{x^*}^{\infty} n(x,t)dx$. It follows from Equation (1) that:

$$\frac{dN}{dt} = \psi(x^*,s)n(x^*,t) - \int_{x^*}^{\infty}\int_{x^*}^{\infty} b(x',x)n(x',t)n(x,t)dxdx'$$

We have studied the evolution of $n(x,t)$ by numerical simulation using techniques described in detail elsewhere. These simulations use the continuum diffusive growth laws:

$$\psi(x,s) = 4\pi RDC_V(s - exp(K/R))$$

where $x = (4\pi/3)R^3 d$, D is the binary diffusion coefficient of monomer in host gas and K is the Kelvin coefficient for the dependence of vapor pressure on droplet curvature. d is the mass density of a particle with radius R. The rate coefficient for coalescence is that appropriate to the continuum Brownian process:

$$b(x',x) = (2kT/3\mu)(x^{1/3} + x'^{1/3})\left(\frac{1}{x^{1/3}} + \frac{1}{x'^{1/3}}\right)$$

where kT is the thermal energy and μ is the viscosity coefficient of the host gas.

Equation (1) may be put into nondimensional form through the substitutions: $\rho = x/\bar{x}_o$, $\tau = 4\pi DC_V(3/4\pi d\bar{x}_o^2)^{1/3}t$, $\phi = \bar{x}_o n(x,t)/N_o$. $\bar{x}_o$ is the initial mean particle mass and N_o the initial number concentration of particles. In these variables, Equation (1) becomes:

$$\frac{\partial\phi(\rho,\tau)}{\partial\tau} + \frac{\partial}{\partial\rho}[\rho^{1/3}(s - \exp(K'/\rho^{1/3}))\phi(\rho,\tau)] =$$

$$\alpha\int_{\rho^*}^{\rho/2} b_1(\rho-\rho',\rho')\phi(\rho-\rho',\tau)\phi(\rho',\tau)d\rho' - \phi(\rho,\tau)\int_{\rho^*}^{\infty} b_1(\rho',\rho)\phi(\rho',\tau)d\rho' \quad (3)$$

where

$$\alpha = kTN_o\bar{x}_o^{2/3}/3\pi\mu DC_v(3/4\pi d)^{1/3}$$

The coefficient α is a measure of the relative rates of coalescence to condendation/evaporation. In Equation (3), for $\alpha \to \infty$, the coalescence process becomes dominant and for $\alpha \to 0$, only the condensation/evaporation process remains with results previously described.[3,12]

A question addressed by others[8,16] using scaling arguments is which mechanism - condensation/evaporation or coalescence - will be dominant for intermediate values of α. The conclusion from these analyses is that asymptotically coalescence will dominate at higher particle mass concentrations - that is for larger values of α. We show here that this is not the case and that apparently coalescence can only be dominant asymptotically for the growth laws studied for vanishingly small rates of the condensation/evaporation process. This is illustrated by presenting a result from numerical solution of Equations (1) and (2) for the stated growth laws. Parameters used yield: $\alpha = 0.31$, $\tau = 5.3 \times 10^3 t$, $s(t=0) = 10$, $K/\bar{R}_o = 1.0$.

Figure (1) shows the evolution in time of the moments N, M_1 of the distribution together with variation with time of the interfacial diameter, $D_p^* : D_p^* = 2K/\ln S$. After an initial induction period[11,12] it can be noted that $N(t) \sim t^{-1}$. This dependence is the same predicted for asymptotic continuum diffusive growth by the condensation/evaporation process according to the Lifshitz-Slyozov (LS) theory.[3] It is also the same dependence found both numerically and by similarity theory[17] for the continuum Brownian coalescence process. The increase of D_p^* with time follows the law: $D_p^* \sim t^{1/3}$, also the same predicted by the LS theory for only the condensation/evaporation process. Interestingly, even substantial coalescence rates do not alter significantly this result from condensation/evaporation similarity theory.

Since, separately, condensation/evaporation and coalescence both give the result, $N(t) \sim t^{-1}$, for the continuum diffusive cases, scaling arguments are insufficient to establish which of the two mechanisms is dominant asymptotically.

Figure (2) shows the contribution to the total rate of change of N, dN/dt, by each of the two processes - condensation/evaporation and coalescence. The continuous curve is that for the ratio of coalescence rate of change of N to the total rate and the dashed curve that for the condensation/evaporation process according to Equation (1). Clearly, the contribution of coalescence to the change of N decreases with time while that for condensation/evaporation increases. Therefore previous conjectures on the dominance of either of the two processes appear to be incorrect for these continuum diffusive growth processes.

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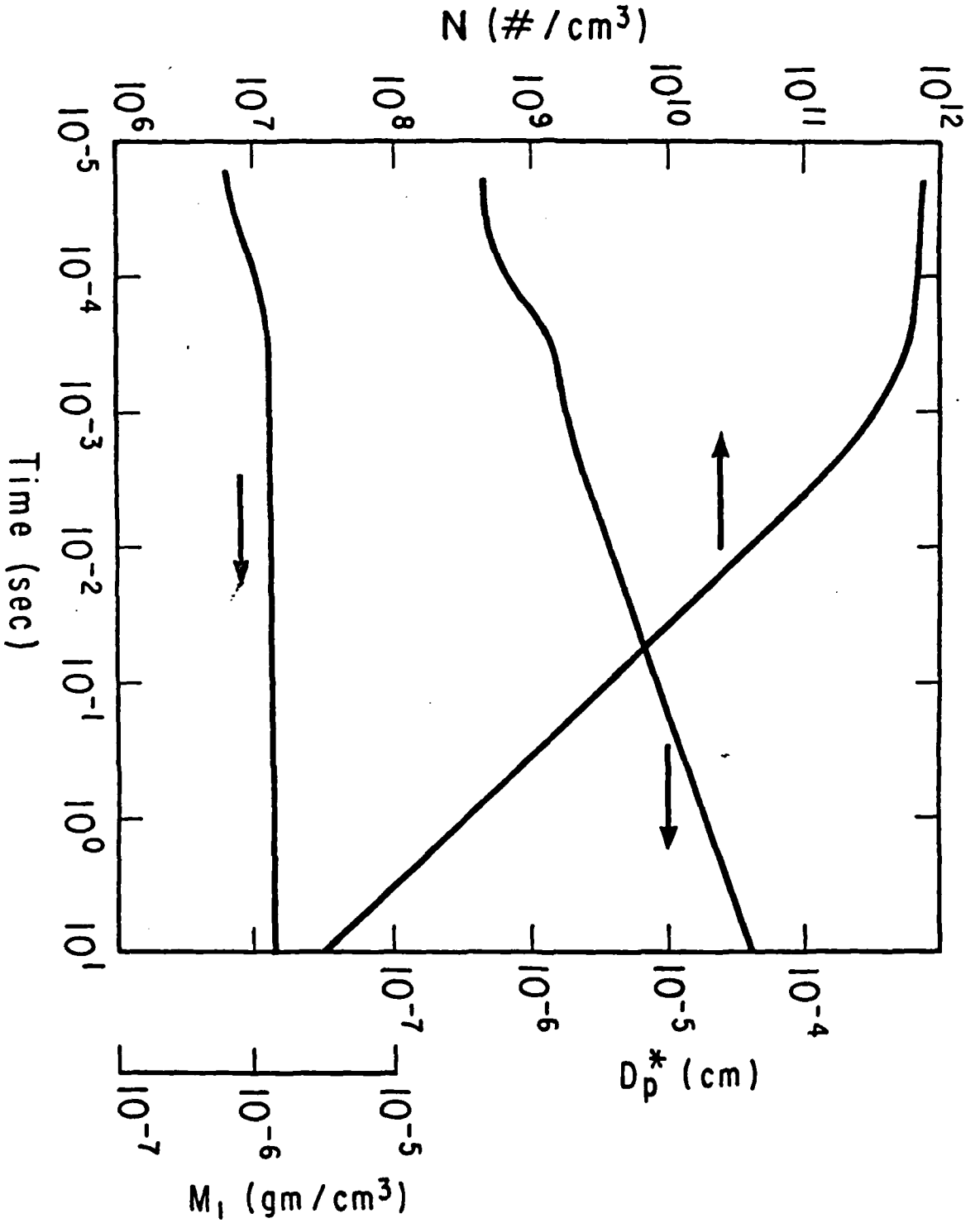
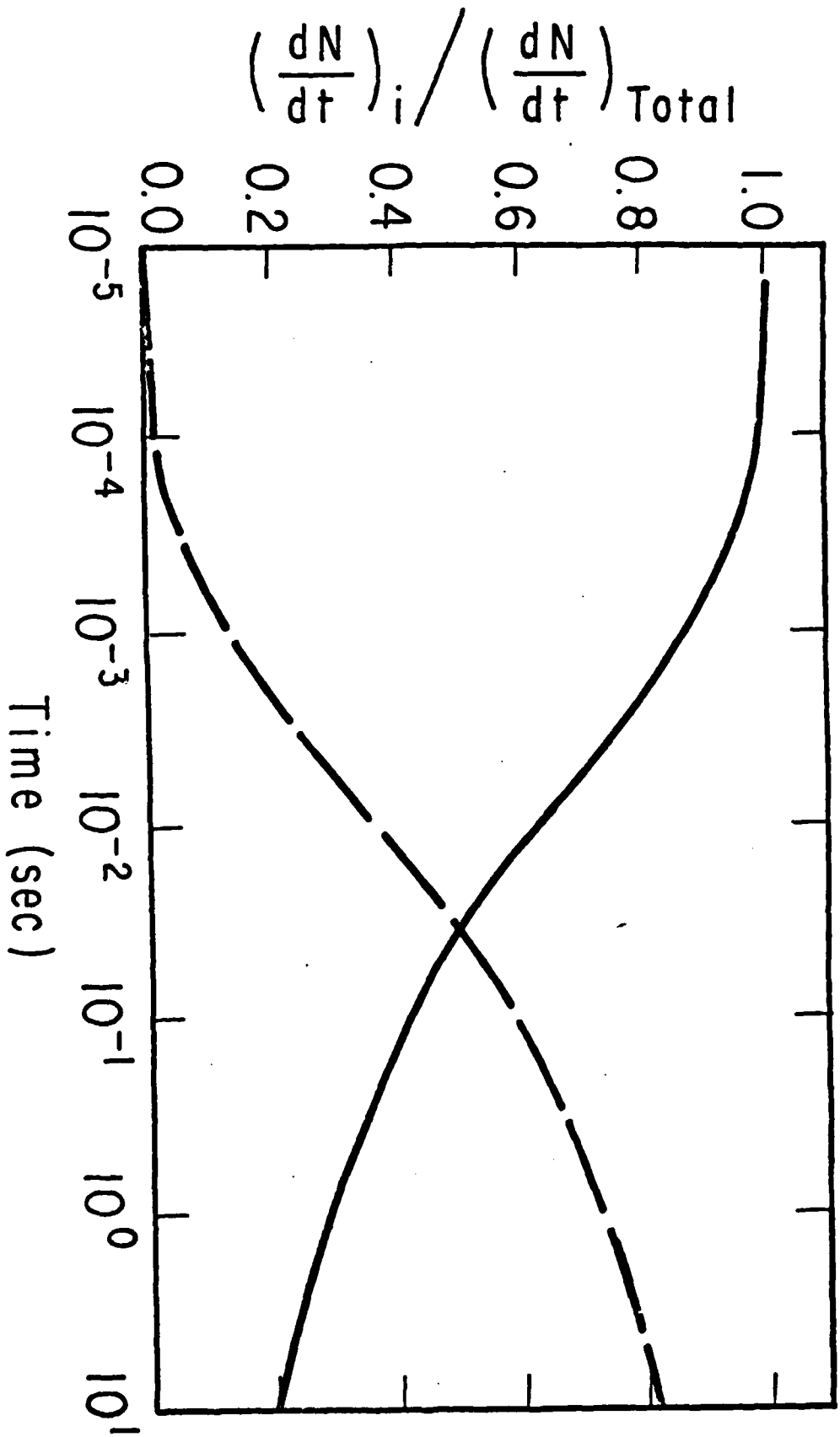
FIGURE CAPTIONS

FIGURE 1. Evolution of total particle number concentration, N, and total particle mass concentration for continuum diffusive growth with simultaneous processes of condensation/evaporation and coalescence. D_p^* is particle diameter at condensation/evaporation interface of particle size distribution.

FIGURE 2. Relative contributions of coalescence (solid curve) and condensation/evaporation (dashed curve) to rate of total time change of particle number concentration, N.


N (# / cm³)
10⁶
10⁷
10⁸
10⁹
10¹⁰
10¹¹
10¹²
Time (sec)
10⁻⁵
10⁻⁴
10⁻³
10⁻²
10⁻¹
10⁰
10¹
D_p* (cm)
10⁻⁷
10⁻⁶
10⁻⁵
10⁻⁴
M_I (gm / cm³)
10⁻⁷
10⁻⁶
10⁻⁵


$\left(\frac{dN}{dt}\right)_i / \left(\frac{dN}{dt}\right)_{Total}$
0.0
0.2
0.4
0.6
0.8
1.0
10^{-5}
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}
Time (sec)

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